# Interaction of Surfactants with Shear Flows and Surface Waves

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## LONG-TERM GOALS

The rapid increase in computing power in the past two decades has made it possible to numerically solve the exact equations of fluid motion over a moderate range of scales in time and space. And although the equations of motion for the flow in the air and in the sea, namely the Navier-Stokes equations, are well known, the boundary conditions for these equations are generally unknown at a surfactant-influenced gas/liquid interface such as the ocean surface. This is because the boundary conditions for these equations are functions of the intrinsic viscoelastic properties of the interface, general techniques for their measurements are not yet available.

This problem is highly nonlinear, since the viscoelastic properties depend on the composition of the interface which varies in both time and space due to interfacial motion resulting from, e.g., wind, water currents and waves. Once these intrinsic properties are determined, then the (numerical) solution of the Navier-Stokes equations can fully predict the motion of the interface and its manifestations, including wave generation and dampening, as well as mass, momentum and energy transport across the interface.

The long-term goals of this project are to develop the necessary instrumentation and techniques to noninvasively determine the dynamic (i.e. viscoelastic) properties of the ocean surface, as described by mechanics-based constitutive relations, and to relate these intrinsic properties to the apparent properties of the interface as interpreted by the usual phenomenological approach of wave-dampening. Ultimately, the intrinsic properties will be utilized in predictive computer models for the surfactant-influenced ocean surface.

## SCIENTIFIC OBJECTIVES

The scientific objective is to develop the instrumentation for in situ ocean measurements of the fully-resolved flow field, including the velocity field at and near the interface along with the curvature of the surface, and the thermodynamic state of the interface, namely, the surfactant composition and concentration. Only through simultaneous measurement of the velocity field at the surface and the surface composition, can the intrinsic viscosities of the interface be obtained. The information on the kinematics of the surface when combined with the thermodynamic state of the interface, measured through nonlinear optical probing of the surface (e.g. through SHG or SFG, in collaboration with Prof. Korenowski of RPI's Department of Chemistry), can completely describe the viscoelastic properties of the interface.

The present research effort is focused on measuring the interfacial properties that have a significant influence on the dynamics of c-g wave dampening. These properties arise from the

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description of the surface through the general constitutive relation for a Newtonian interface, namely the Boussinesq-Scriven surface model. This surface model involves two dissipative thermodynamic properties, namely, interfacial dilatational viscosity ($\kappa^s$) and interfacial shear viscosity ($\mu^s$), along with the thermodynamic (i.e. equilibrium or static) surface tension ($\sigma$). Furthermore, all of these properties ($\kappa^s$, $\mu^s$ and $\sigma$) are functions of the thermodynamic state of the interface, i.e. not only does $\sigma$ vary with surfactant concentration c (the well known elastic effect), but $\kappa^s$ and $\mu^s$ also vary with the amount of surfactant adsorbed to the surface. Thus, in addition to these surface properties, the transport kinetics must also be understood to permit the modeling of the interaction between the surfactant in the bulk and on the surface. There are three properties that control the transport of surfactant to and from the interface, namely, the equilibrium ratio of bulk and interfacial concentration (K), interfacial diffusivity ($D^s$), and bulk diffusivity (D). With the exception of $\mu^s$, direct measurements of the dynamic properties have not been feasible until recently. In the present approach, these quantities are found from fully resolved velocity and surfactant concentration measurements utilized along with the tangential-component(s) of the stress boundary condition.

It should be emphasized here, that although there are techniques already described in the literature for the measurement of each of the quantities listed above, consistent measurements have not yet been obtained for several of these properties; in particular, the surface dilatational viscosity ($\kappa^s$). Measurements of $\kappa^s$ for a given surfactant obtained through two different techniques (the Maximum Bubble Pressure Method and the Longitudinal Wave Method) that appear in the literature, differ form one another by *five orders of magnitudes* (see Edwards, Brenner & Wasan, Interfacial Transport Processes & Rheology, 1991)

## APPROACH

The present effort was multifaceted and involved several different experimental projects (in the laboratory) and was augmented through a collaborative theoretical project. The common theme among these tasks was to quantify the surface dilatational and surface shear viscosities in various flow fields involving a water surface covered by a surfactant monolayer.

## WORK COMPLETED

The tasks that were supported (at least in part) by this grant were in the following projects:

I.   **Determination of the sum ($\cdot$ $^S$ + $\cdot$ $^S$) through novel measurements of the velocity field and surfactant concentration on the interface during the interaction of a vortex pair with the free surface**

In this effort, some fundamental fluid dynamics issues as well as many instrumentation issues were addressed through a flow field that has been used in the past for the development of noninvasive measurements at the air-water interface. Using a new 100 Hz Nd:YAG laser system ("Infinity" model from Coherent) from Prof. Korenowski's laboratory, multipoint SHG (second-harmonic generation) measurements were performed, while the flow field in the water was measured with the DPIV (digital particle image velocimetry) technique. The feasibility of several different experimental techniques were examined, including the possibility of directly measuring the surfactant concentration gradient, and direct determination of surface curvature from images of

seeded flow fields obtained for DPIV. This effort lead to the development of Boundary-Fitted DPIV technique, described in the Results Section.

**II.  Measurements of surface shear viscosity ($\mu^s$) of insoluble monolayer films through the classical technique of deep-channel surface viscometer**

A new deep-channel surface viscometer was constructed of stainless steel and used for measurements of surface shear viscosity ($\mu^s$) using the classical linear theory. To facilitate this project, a computer-controlled Langmuir trough was also designed and constructed. The trough was constructed of Teflon. The equation of state ($\sigma$ vs. c) for different insoluble monolayer systems were measured in the Langmuir trough and the information was used to determine the state of the interface in the deep-channel surface viscometer measurements.

**III.  Simultaneous measurements of surfactant concentration (via scanning SHG) and surface strain field (via scanning LDV) on a quasi-steady surfactant film behind a Reynolds ridge**

A new flow loop was specifically designed and constructed to study the free surface on a steady-state, uniformly flowing stream where a surface-piercing barrier had trapped a surfactant film. Aside from the fundamental interest, this flow provided a benchmark for the development and testing of Prof. Korenowski's areal SHG system. The viscous shear stress acting on the interface, determined from the rate of strain on the fluid measured by laser-Doppler velocimetry (LDV), and surface tension gradient (due to surface elasticity) as determined by SHG, were used to quantify the relative importance of surface viscosities and surface elasticity.

**IV.  IV Measurements of the surface deformation on c-g waves using the Boundary-Fitted DPIV technique**

A new wave tank system was designed and constructed to permit optical measurements using the DPIV technique. The original wave tank in our laboratory, which was only 60 cm in length (25 cm wide, 15 cm deep), utilized a plunging wedge driven by audio speakers to generate the waves. That system was only capable of producing relatively high frequency waves, namely, larger than 25 Hz. With a length of 3 m, the new wave tank system is significantly longer than the old one. Only surface-inactive material (glass, Teflon, and stainless steel) were utilized in the construction of the facility which is filled with doubly-distilled water from a glass distiller. A paddle-type wave generator is utilized in the new facility. A computer-controlled stepper motor provides a nearly sinusoidal motion of the paddle (the standard deviation from a true sine wave is less than 1% of the peak). Stable, two-dimensional surface waves with initial amplitude of more than 1 mm have been generated with frequency ranging from 1 to 20 Hz in the new facility. Wave reflection from the end of the tank was minimized using a glass (i.e. zero contact angle surface) beach with a shallow inclination. A surface-piercing Teflon barrier with side-wall seals was used for sweeping any residual surfactants to the end of the tank where is was drawn up using surface suction. Furthermore, a permanent surface drain ensured a very clean initial condition.

The maximum wave height can be measured independently in this facility with a mechanical surface probe to an accuracy of ±10 microns to check the accuracy of the results of surface location measurements via the Boundary-Fitted DPIV technique, described in item-I in the Results Section, below.

The boundary-fitted DPIV technique was tested in this facility with 3-10 Hz surface waves. The experiments were conducted with a clean free surface; contaminated surface cases will be conducted in the future by the student supported through the accompanying AASERT grant (June 1, 1997 - May 31, 2000).

## V. Navier-Stokes numerical simulations of a deep-channel viscometer flow which is driven into the nonlinear regime

Theoretical and computational studies have been undertaken to complement the experimental effort on the hydrodynamic coupling of the bulk flow with contaminated interfaces. In a collaborative effort with Prof. Juan Lopez of Arizona State University, Department of Mathematics, the flow in a deep-channel viscometer was simulated to examine the effect of the surface shear and dilatational viscosities as well as surface elasticity on the fluid flow field. Experiments are presently being conducted to support this effort. The present measurements are limited to visualizations of particle trajectories on the surface. Measurements using the boundary-fitted DPIV technique, as well as surfactant surface concentration measurements via areal SHG technique, in collaboration with Prof. Korenowski, are planned for the future.

## RESULTS

The fundamental results obtained in each of the 5 projects outlined above are presented here:

I. A new approach for the determination of surface viscosities has been developed. This approach utilizes surfactant concentration data from nonlinear optics measurements (work performed in collaboration with Prof. Korenowski) along with measurements of the velocity field. In this way, every term in the stress boundary condition is measured and the numerical value of the sum of the surface dilatational and shear viscosities is determined.

The scientific results of some of the collaborative effort between our group and Prof. Korenowski's has been published. In a paper published last year in the *Experiments in Fluids* (Hirsa et al. Vol. 22, No. 3, pp. 239-248 [1997]), the technique for the simultaneous measurement of the surfactant concentration (via SHG) and velocity field (via DPIV) was described. The analysis presented in that paper showed, for the first time, that quantitative information on the subsurface flow field can be directly obtained from nonlinear optics probing of the surface composition.

The technique for the direct measurement of the sum of the intrinsic surface viscosities ($\kappa^s + \mu^s$) was also published last year in the journal *Langmuir* (Hirsa et al. Vol. 13, No. 14, pp. 3813-3822 [1997]). The theory behind our technique along with examples of fully-resolved measurements of the free surface boundary layer were included in that paper. In its present form, the technique for the direct measurement of the surface viscosity is applicable to insoluble system. The extension of the technique to soluble systems, and ultimately to natural ocean films is the central thrust of the present effort and is expected to become feasible in the near future, as the development of the areal SHG system is nearing its completion. The theoretical foundations for the extension of this approach to the determination of surface viscosities, without the usual limitations of constant property formulation, is described in Section-V, below.

Recently, a novel technique referred to as Boundary-Fitted DPIV, has been developed and successfully tested which provides the instantaneous shape of the air-water interface as well as the

velocity field. The data obtained through this technique significantly improves the accuracy of surface viscosities computed from the velocity data. This technique is a hybrid form of DPIV in which particle images are first utilized to map the location of the surface through a digital cross-correlation method. This is possible because of the total internal reflection that occurs when viewing submerged objects from a shallow angle below the surface. Surface elevations of up to a few millimeters at any x-position can thus be determined to an accuracy of order 10 microns. The primary limitation of the Boundary-Fitted DPIV technique is the minimum radius of curvature that it can resolve, i.e. the method works well for locating and measuring surfaces with arbitrarily large radii of curvature (with the maximum radius corresponding to a flat surface), but the minimum radius of curvature measurable is larger than the correlation window size (of order 100 microns) used in the technique.

This new technique, which in its present form provides a one-dimensional mapping of the surface, has been incorporated into our DPIV software to provide the velocity field in boundary-fitted coordinates, i.e. measurements on grids fitted to the (deforming) air-water interface. The boundary-fitted measurements are particularly important because of the large gradients that exist in the velocity within the free-surface boundary layer. The preliminary results from the Boundary-Fitted DPIV measurements were presented at the APS meeting last November (see item #3 in the publications list, below). A thesis (Gayton 1997) has been written on this technique and we are presently preparing a manuscript to submit to *Experiments in Fluids* .

The application of this technique to c-g surface waves has also been initiated. The preliminary results appear to be successful and are described in Section-IV, below.

II.    Since the technique described in Section-I provides the sum of the surface shear and the surface dilatational viscosities (see Hirsa et al. *Langmuir*, 1997), a separate measurement of the surface shear viscosity ($\mu^s$) was needed to determine the surface dilatational viscosity ($\kappa^s$). Although measurement of $\mu^s$ is not as difficult as that of $\kappa^s$, it is nevertheless a formidable task to make repeatable and consistent measurements of $\mu^s$. The technique of deep-channel surface viscometry was selected for measurement of $\mu^s$, as it is commonly agreed to be the most sensitive technique presently available.

A high precision, deep-channel surface viscometer was designed and constructed for the present project. The viscometer was constructed of stainless steel to reduce the possibility of surface contamination. Recently, a set of measurements of $\mu^s$ were made for hemicyanine monolayers (insoluble) spread on water. Measurements for hemicyanine concentrations ranging from 0.2 to 1.1 (mg/m$^2$) have been completed. The data from these measurements are being utilized in a computational model (see item #16 in the publications list, below), and the viscosity results will be reported at the APS meeting in November (see item #18 in the publications list, below). Measurements of $\mu^s$ for two other surfactant systems, stearic acid, which is a relatively viscous monolayer, and oleyl alcohol, which is a monolayer with a relatively low viscosity, are presently underway.

III.    Noninvasive techniques were utilized for the first time for determining the instantaneous subsurface viscous shear stress (via scanning laser-Doppler velocimetry, LDV) and the spatial gradient of surfactant concentration field (via scanning, point-wise second-harmonic generation, SHG, system and an areal SHG system) for a quasi-steady flow at an air/water interface. The measurements were made on a surfactant film trapped by a surface barrier piercing a uniform

flow. A Reynolds ridge marks the leading edge of the boundary layer formed beneath the surfactant film. Interestingly, the measured shear stress acting on the surfactant film, as obtained from the LDV data, was found to be *greater* than the shear stress for a solid wall. This may be because of the presence of secondary flow on the surfactant-covered surface examined, in particular, the reverse surface flow observed along the tunnel centerline. Some of the results from this study were reported at the APS meeting last November (see item # 12 in the publications list, below).

The surface tension gradient, computed from the SHG data, was compared to the viscous shear stress, obtained from the LDV data. Preliminary analysis shows that the effect of the intrinsic surface viscosities ($\kappa^s$ and $\mu^s$) is not negligible compared to the elastic term, $\frac{\partial \sigma}{\partial x}$. A Masters thesis (Jin 1998) has been written on this work and we are planning additional collaborative studies with Prof. Korenowski's group.

IV.    The noninvasive technique of Boundary-Fitted DPIV, developed in our laboratory for project I, has successfully been applied to the measurement of small-amplitude (linear or weakly nonlinear) surface waves. Preliminary measurements taken with the wave-maker driven at 3 Hz, have confirmed that the frequency of the waves can be readily obtained from the Boundary-Fitted DPIV technique; experiments to test whether or not the wave amplitude obtained from the images agrees with the mechanical wave gauge measurements have yet to be performed.

V.    The final area of research directly related to this grant is the theoretical and computational study of free-surface flows in the presence of surfactants. So far, we have published one paper in this area (Lopez & Hirsa, Journal of Colloid and Interface Science, Vol. 206, No. 1, pp 231-239 [1998]). In this paper, we presented the theoretical foundation, based on the Boussinesq-Scriven surface model without the usual simplification of constant viscosities, for an experimental technique to directly measure the surface shear ($\mu^s$) and dilatational ($\kappa^s$) viscosities for a Newtonian interface as functions of the surfactant surface concentration.


**IMPACT FOR SCIENCE**

We have shown for the first time that the intrinsic viscoelastic properties of surfactant-influenced surfaces may be measured directly through optical techniques. Preliminary results have been obtained that show that the intrinsic properties can be utilized in numerical simulations of the axisymmetric Navier-Stokes equations. These results provide the foundations to predicting the fully three-dimensional and unsteady behavior of fluid flow on each side of the air-sea interface.


**TRANSITIONS ACCOMPLISHED AND EXPECTED**

We expect to develop the instrumentation and techniques to perform Boundary-Fitted DPIV measurements of the velocity field in the free surface boundary layer beneath ambient waves on the ocean in the next few years; this capability will complement the nonlinear optical measurements of the surface which has already been proven in situ.

## RELATIONSHIP TO OTHER PROJECTS

The present research project is closely linked to the research in nonlinear optical probing of the ocean surface, conducted by Prof. Korenowski in the Chemistry Department at RPI. Our synergistic approach is designed in such a way that their efforts will continue to provide us with the tools needed for measurements of the viscoelastic interfacial properties in the laboratory and ultimately in situ, and our flow facilities and measurement capabilities provide the benchmark for the development and testing of their surface probes prior to their implementation for in situ studies.

## RECENT PUBLICATIONS FROM ONR WORK

Amir Hirsa

1) 96-P    Logory, L.M., Hirsa, A. and Anthony, D.G., "Interaction of Wake Turbulence with a Free Surface," *Physics of Fluids* **8** (3), 805-815 (1996).

2) 96-IC    Hirsa, A., "Interaction of Turbulence with an Air-Water Interface and the Effects of Surfactants," invited presentation at the State University of New York, Buffalo, NY (Apr. 1996).

3) 96-C    Gayton, J.D., Hirsa, A., Korenowski, G.M. and Logory, L.M., "Determination of Surface Viscosities by Surfactant Concentration and Velocity Field Measurements," presented at 49th Annual Meeting, APS, Div. of Fluid Mechanics, Syracuse, NY (Nov. 1996). Abstract in the Bulletin of APS, Vol. 41, No. 9, 1746 (1996).

4) 96-C    Hirsa, A., Lopez, L.M. and Kim, S., "Interaction of a Columnar Vortex with a Solid Wall," presented at 49th Annual Meeting, APS, Div. of Fluid Mechanics, Syracuse, NY (Nov. 1996). Abstract in the Bulletin of APS, Vol. 41, No. 9, 1797 (1996).

5) 97-P    Hirsa, A., Korenowski, G.M., Logory, L.M. and Judd, C.D., "Velocity Field and Surfactant Concentration Measurement Techniques for Free-Surface Flows," *Experiments in Fluids* **22** (3), 239-248 (1997).

6) 97-P    Hirsa, A., Korenowski, G.M., Logory, L.M. and Judd, C.D., "Determination of Surface Viscosities by Surfactant Concentration and Velocity Field Measurements for an Insoluble Monolayer," *Langmuir* **13** (14), 3813-3822 (1997).

7) 97-IC    Hirsa, A., "Hydrodynamic Coupling of Viscoelastic Gas/Liquid Interfaces," invited presentation at the Department of Mathematics, Pennsylvania State University, State College, PA (Apr. 1997).

8) 97-IC    Hirsa, A., "Fluid Dynamics of Contaminated Gas/Liquid Interfaces," invited presentation at the Department of Environmental and Energy Engineering, Rensselaer Polytechnic Institute, Troy, NY (Nov. 1997).

9) 97-C    Hirsa, A., Gayton, G.D., Korenowski, G.M., Lopez, J.M. and Chen, S., "Hydrodynamic Coupling of Surfactant-Influenced Gas/Liquid Interfaces," paper number 97-2057 presented at the 28th AIAA Fluid Dynamics Conference, Snowmass, CO (June 1997).

10)  97-C  Mortzheim, J.P., Gayton, J.D. and Hirsa, A., "Free Surface Velocity and Elevation Measurement Techniques as Applied to the Interaction of a Vortex Pair with a Contaminated Interface," presented at 50th Annual Meeting of the American Physical Society, Div. of Fluid Mechanics, San Francisco, CA (Nov. 1997). Abstract in the Bulletin of APS, Vol. 42, No. 11, 2163 (1997).

11)  97-C  Hirsa, A., Lopez, J.M. and Mortzheim, J.P., "Direct Determination of Surface Dilatational Viscosity," presented at 50th Annual Meeting, American Physical Society, Div. of Fluid Mechanics, San Francisco, CA (Nov. 1997). Abstract in the Bulletin of APS, Vol. 42, No. 11, 2176 (1997).

12)  97-C  Korenowski, G.M., Hirsa, A., Van Wagenen, E. and Jin, H.J., "Optical Technique for the Areal Measurement fo Surfactant Concentration as Applied to a Monolayer Downstream of a Reynolds Ridge," presented at the 50th Annual Meeting, American Physical Society, Div. of Fluid Mechanics, San Francisco, CA (Nov. 1997). Abstract in the Bulletin of APS, Vol. 42, No. 11, 2224 (1997).

13)  98-P  Lopez, J.M. and Hirsa, A., "Direct Determination of the Dependence of the Surface Shear and Dilatational Viscosities on the Thermodynamic State of the Interface: Theoretical Foundations," *Journal of Colloid and Interface Science* **206** (1), 231-239 (1998).

14)  98-PS  Hirsa, A., Lopez, J.M. and Kim, S., "Evolution of an Initially Columnar Vortex Terminating Normal to a No-slip Wall," *Experiments in Fluids* (submitted).

15)  98-PI  Hirsa, A. and Gayton, J.D., "Velocity and Surface Elevation Measurement Technique for Free-Surface Boundary Layers," *Experiments in Fluids* (in preparation).

16)  98-PI  Hirsa, A. and Lopez, L.M., "Modeling of Surface Dilatational Viscosity through High-Reynolds Number Deep-Channel Viscometer Measurements and Navier-Stokes Computations," *Journal of Colloid and Interface Science* (in preparation).

17)  98-C  Van Wagenen, E., Korenowski, G.M., Jin, H.J., Hirsa, A. and Saylor, J.R., "Imaging a Surfactant Concentration Gradient Downstream of a Reynolds Ridge," presented at the American Chemical Society Meeting, University Park, PA (June 1998).

18)  98-C  Hirsa, A., Lopez, J.M., Korenowski, G.M., Ringuette, M.R., and Joshi, A., "Measurements in a High Reynolds Number Deep-Channel Viscometer," presented at the 51st Annual Meeting of the American Physical Society, Div. of Fluid Mechanics, Philadelphia, PA (Nov. 1998). Abstract in the Bulletin of APS.

19)  98-C  Lopez, J.M. and Hirsa, A., "Direct Determination of Surface Shear and Dilatational Viscosities," presented at the 51st Annual Meeting of the American Physical Society, Div. of Fluid Mechanics, Philadelphia, PA (Nov. 1998). Abstract in the Bulletin of APS.